Evaluating Ozone Precursor Emission Inventories

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"Experience with the 1990 State Implementation Plan (SIP) base year (emissions) inventories brought to light deficiencies and inconsistencies in the inventory process now being used. In addition, the current leeway in selecting these processes has resulted in data sets of unknown quality and varying degrees of completeness." - *Emission Inventory Improvement Program, 1997*

Ozone precursor emission inventory development is critical because the inventory is used:

- To support air quality modeling (i.e., model input)

- To support exposure modeling and health assessments

- To analyze emission control costs

- To develop regulatory control strategies

> "The Clean Air Act requires state and local air quality agencies to develop complete and accurate inventories as an integral part of their air quality management responsibilities. These air emission inventories are used to evaluate air quality, track emissions reduction levels, and set policy on a national and regional scale...." - *Emission Inventory Improvement Program, 1997*

Introduction (2 of 2)

- Important ozone precursors include:
  - oxides of nitrogen (NO_x)
  - volatile organic compounds (VOC)
  - carbon monoxide (CO)
- Sources of these precursors include:
  - NO_x emissions primarily from anthropogenic sources involving fossil fuel combustion (i.e., motor vehicle exhaust and coal combustion).
  - NO_x emissions naturally occurring (in small quantities) from microbial actions in soils following the application of fertilizer.
  - Anthropogenic (man-made) VOC emissions from a broad range of sources, including mobile (e.g., cars and trucks) and stationary (e.g., evaporation of solvents and gasoline, dry-cleaning, and auto repair shops).
  - Biogenic (natural) VOC emissions released by trees and plants.
  - CO emissions from combustion processes (i.e., stationary and mobile combustion sources).
- PAMS data are useful for evaluating emission inventory estimates of total VOC, individually speciated compounds, NO_x, and VOC/NO_x ratios.

The Importance of Emission Inventory Evaluation

Why bother evaluating emissions data?

- Emission inventory development is a complex process that involves estimating and compiling emissions activity data from hundreds of point, area, and mobile sources in a given region. Because of the complexities involved in developing emission inventories and the implications of errors in the inventory on air quality model performance and control strategy assessment, it is important to evaluate the accuracy and representativeness of any inventory that is intended for use in air quality modeling.

What tools are available for assessing emissions data?

- There are several techniques used to evaluate emissions data including: "common sense" review of the data; source-receptor methods such as the chemical mass balance model; bottom-up evaluations that begin with emissions activity data and estimate the corresponding emissions; and top-down evaluations that compare emission estimates to ambient air quality data. Each evaluation method exhibits strengths and limitations.

- Based on the results of the emissions evaluation, recommendations can be made on possible improvements to the emission inventory. Local agencies responsible for developing the inventory can then make revisions to the inventory data prior to air quality modeling.

- Estimating ozone precursor emissions is a complex process involving many parameters.

- Uncertainties in emissions inventory estimates could range from about 10% for well-defined sources to an order of magnitude for widespread and sporadic sources

- The general equation for estimating emissions is:

$$E = A \times EF \times (1-ER/100)$$

Where:

E = emissions   EF = emission factor

A = activity   ER = overall emission reduction efficiency (%)

Spatial Allocation of Emissions Activities

- Emission sources are spatially allocated to a region using the actual locations of the emissions sources and/or using spatial surrogate data which are physical parameters that can be associated with emissions activities (e.g., acres of farmland might be the surrogate for emissions from farming operations).

Temporal Allocation of Emissions Activities

- Emission sources are temporally allocated by assigning a temporal profile, a distribution of emissions activity over a 24-hr period, to each source category.

Chemical Speciation of Emission Sources

- In order to disaggregate VOC emissions into individual chemical species, each emission source category is assigned a speciation profile which provides a detailed chemical breakdown of the individual chemical species emitted from that source.

- Several sources of VOC speciation data currently exist including:

    – EPA's SPECIATE: *http://www.epa.gov/ttn/chief/software.html#speciate*

    – Desert Research Institute (DRI): *http://www.dri.edu*

    – North American Front Range Air Quality Study: *http://www.nfraqs.colostate.edu/index2.htm*

Emission Inventory Evaluation Tools and Methods

Use of mathematical techniques to evaluate emission estimates:

- Engineering judgment approach

- Bottom-up emissions evaluation

Use of ambient air quality data to evaluate and reconcile emission estimates:

- Multivariate techniques and source-receptor modeling

- Top-down emissions evaluation

Using Engineering Judgment to Evaluate Emission Estimates (1 of 2)

- Begin with knowledge of the region for which the emission inventory was developed (i.e., likely emission sources, population, demographic characteristics).

- Review major sources of emissions and perform per-capita checks combined with conventional wisdom to evaluate emissions data.

Strengths

- Provides a quick and inexpensive method to quality control emission estimates.
- Does not require extensive data.
- Can quickly identify gross errors in emissions data.

Limitations

- Can identify errors in emissions data but gives no insight as to where errors emanate.

Summer emission inventories often report significant VOC and/or NO_x emissions from seasonal sources such as snow removal equipment and snowmobiles.

Snowmobile emissions in the summer?

Snow removal equipment and snowmobiles are contributors to VOC and NO_x emissions in the wintertime. Emission contributions from this source should be low in the summer months. Taking the time to review seasonal emission inventory data can catch errors like this.

Bottom-up Evaluation of Emissions Activity Data (1 of 3)

- Bottom-up evaluations are a method of assessing emissions data using census information and emissions activity data combined with emission factors to generate independent estimates to compare to existing data.

- This method is most useful when combined with the top-down evaluation when assessing large data sets. Top-down evaluations identify problem categories; bottom-up evaluations are used to investigate the underlying information used to estimate categories.

Strengths

- The emission estimates generated using this methodology can be very accurate if demographic and activity data are accurate.

Limitations

- Extensive data requirements.
- Accuracy of emission factors.
- Accuracy of activity data.
- Time consuming.

Mobile source emissions activity data for anonymous city:

Urban region with a total fleet of 366,699 on-highway motor vehicles.

Emissions data reported that 494 of these vehicles are heavy-duty diesel trucks (HDDTs).

According to these figures, only 0.1% of the vehicle fleet are HDDTs.

In other parts of the country with similar characteristics, HDDTs make up approximately **10 to 20%** of highway vehicles. HDDTs are significant contributors to NO_x; consequently, errors in activity data can lead to errors in emissions estimates.

Adapted from Haste et. al., 1998

VOC emissions from vehicle refueling:

Step 1. Using reported VOC emissions and EPA emission factors, back-calculate fuel dispensed (e.g., 400,000 gal/day).

Step 2. Evaluate gasoline dispensed for reasonableness.

- In County A, each of the 20,000 registered vehicles would need to have been refueled 20 gallons per day.

- Based on reported vehicle miles traveled (1,600,000 miles/day), average fuel economy would be about 4 miles per gallon (80 miles/day ÷ 20 gal/day).

Top-down emissions evaluation is a method of comparing emissions estimates with ambient air quality data. Ambient/emission inventory comparisons are useful for examining the relative composition of emission inventories; they are not useful for verifying absolute pollutant masses unless they are combined with bottom-up evaluations. The top-down method has demonstrated success at reconciling emission estimates of VOC, CO, and NO_x.

> Early morning sampling periods are the most appropriate to use in these evaluations because emissions are generally high, mixing depths are low, winds are usually light, and photochemical reactions are minimized.

Ambient Data Requirements

- Select ambient monitoring sites dominated by fresh urban source emissions (e.g., PAMS Type II).

- Validate and process ambient VOC, NO_x, and CO data.

- Select early morning (e.g., 0700-0900) hourly data.

- Analyze meteorological data to determine the emission areas and elevated point sources that may influence ambient measurements.

Emissions Data Requirements

- Evaluate emissions for the same locations as the ambient monitor.

- Process emissions data to get gridded, hourly emissions (i.e., match species).

- Convert emissions data units to be compatible with ambient data units.

- Existing emissions processing software: SMOKE, EPS 2.0 or 2.5, EMS-95

Analysis Objectives:

- Compare ambient data with emission estimates from different wind quadrants surrounding the monitoring site.

- Compare ambient data with emission estimates with and without elevated point source emissions. The inclusion of elevated point sources will depend on the meteorological conditions (i.e., morning mixing height).

- Compare VOC/NO_x and CO/NO_x ratios for day specific, weekday, and weekend data.

- Compare individual chemical species in the ambient air to chemical species in the emission inventory when speciation data are available.

Wind Quadrant Definitions Used in the Top-down Evaluation
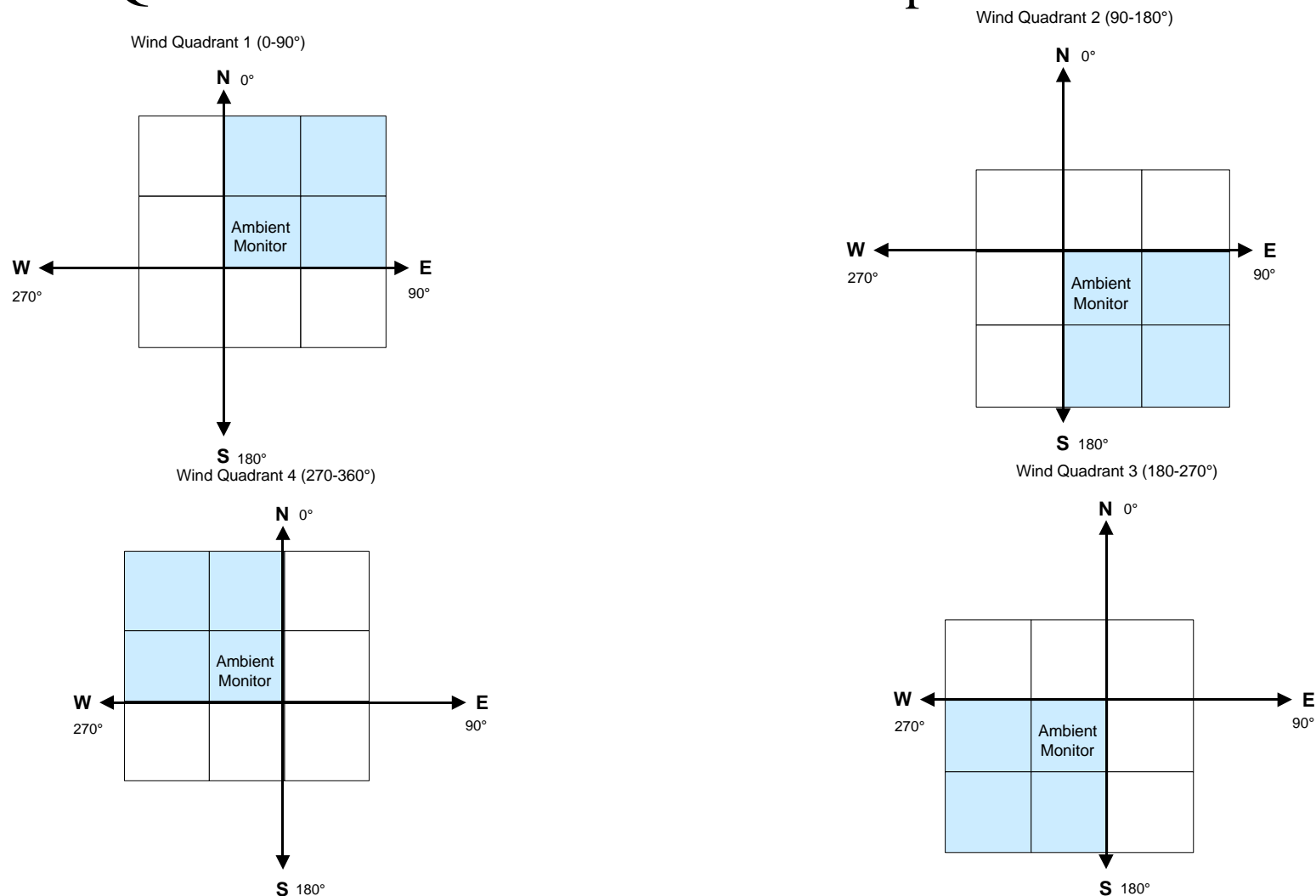
- Compile ambient data.

- Set/apply screening criteria.

- Summarize results.

Site	# Valid Samples[a]	Average TNMOC ppbC[b]	Average ID NMOC ppbC[c]	Average NOx ppb[d]	Average CO ppb
Site 1	18	482	402	64	1268
Site 2	14	467	270	84	1458
Site 3	19	620	404	89	1459

[a] Only valid samples collected from July through September of 1997 (begin hours 0500, 0600, and 0700 PST) meeting all screening criteria were used to calculate the averages.

[b] TNMOC = Total Non-Methane Organic Compounds includes both identified and unidentified compounds. Averages calculated for all valid samples with TNMOC > 100 ppbC.

[c] ID NMOC = Identified Non-Methane Organic Compounds includes only those chemical compounds identified in the ambient air for samples corresponding to TNMOC > 100 ppbC.

[d] NO_x averages calculated for all valid samples with NO_x > 10 ppb to eliminate NO_x values at or near instrument detection limits.

- In processing the emission inventory data, consider only the chemical species capable of being detected by the ambient measurement systems. The adjusted NMOC emissions usually include C2-C10 alkanes, alkenes, alkynes, aromatics, and C1-C3 carbonyls. The emissions of alcohols, ethers, acetates, glycols, esters, formates, organic amines, organic oxides, phenols, organic acids, C3+ carbonyls, terpenes, C11+ hydrocarbons, and halogenated species should usually be excluded in a comparison of the inventory and PAMS data. The emissions of the chemical species capable of being detected by the ambient measurement system usually contribute about 75% of the total emissions in the inventory.

- Convert emissions from mass to molar basis for a valid comparison between ambient data and the emission inventory.

Species Group Emissions Contributions
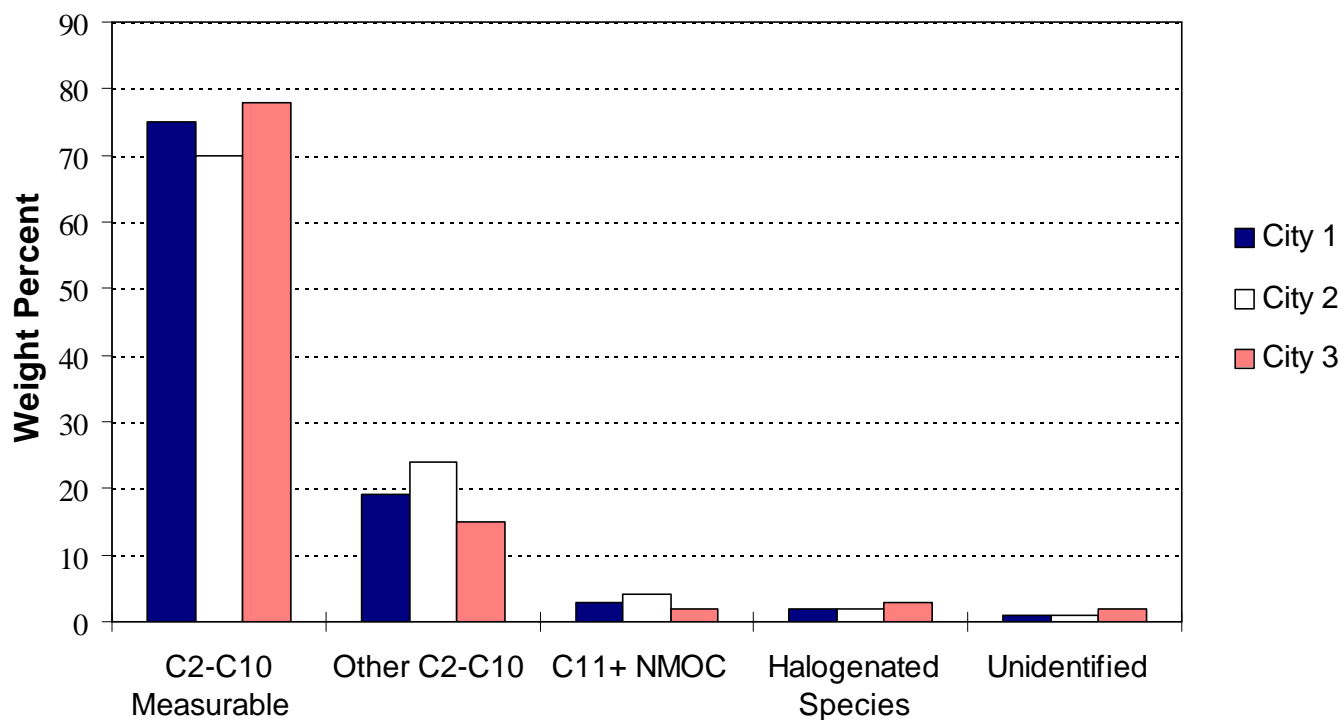
Example plot of morning weight percent contributions of species group emissions for the 9x9 cell (36x36 km) areas surrounding three ambient monitoring sites. The emissions of the chemical species capable of being detected by the ambient measurement system usually contribute about 75 percent of the total emissions in the inventory.

- Prepare tables/plots of VOC/NO$_x$ and CO/NO$_x$ ratios for ambient and emission inventory (with and without elevated point sources).

- Notice the difference in the emission inventory and ambient ratios overall and by wind quadrant. The emission inventory underpredicts the VOC/NO$_x$ ratios observed in ambient data; this may indicate an underestimation of VOC in the emission inventory.
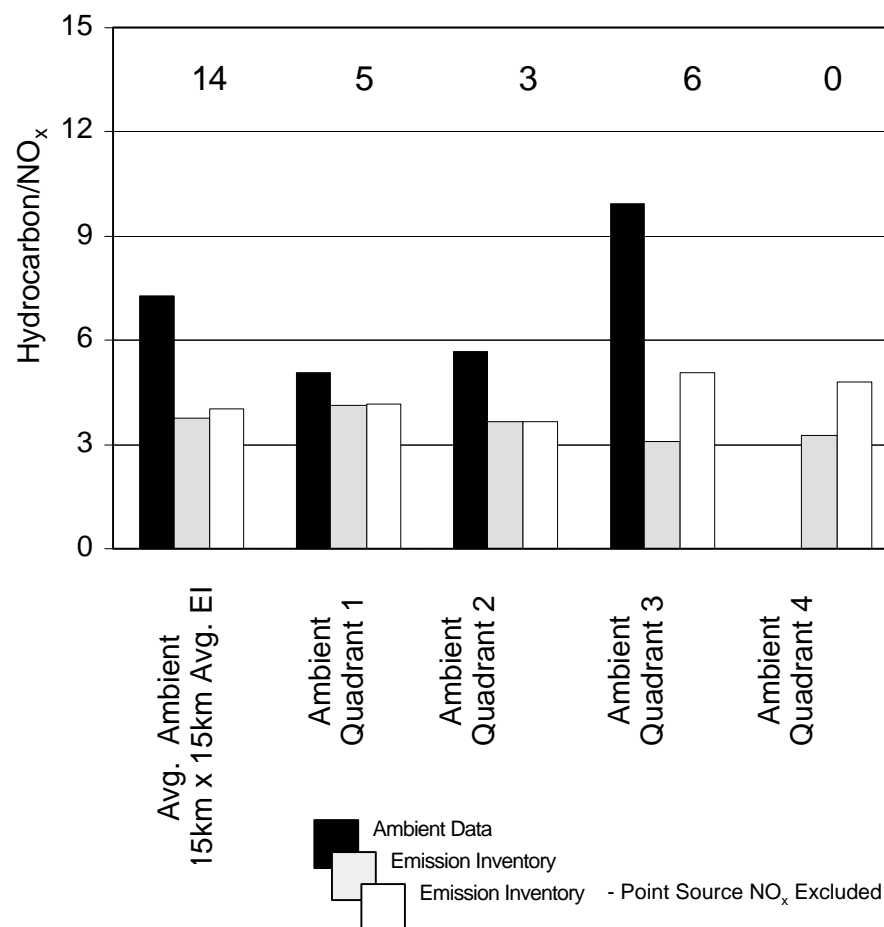
Prepared using Excel. The numbers above the bars represent the number of ambient samples used in the comparison (Chinkin et al., 1999).

- Prepare individual VOC species comparisons.
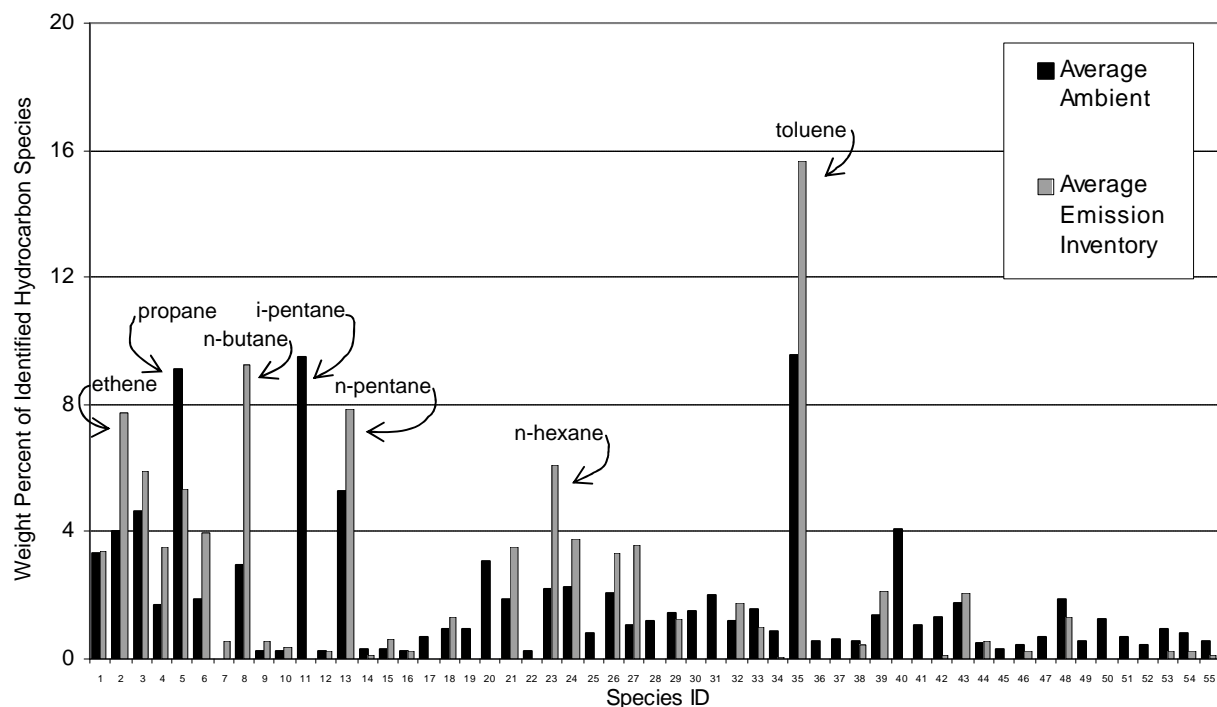
Notice the large differences for some species such as toluene, ethene, and n-butane. These discrepancies indicate the need for improved speciation profiles and/or assignments.

1	acetylene
2	ethylene
3	ethane
4	propylene
5	propane
6	isobutane
7	1-butene
8	n-butane
9	trans-2-butene
10	cis-2-butene
11	isopentane
12	1-pentene
13	n-pentane
14	isoprene
15	trans-2-pentene
16	cis-2-pentene
17	2,2-dimethylbutane
18	cyclopentane
19	2,3-dimethylbutane
20	2-methylpentane
21	3-methylpentane
22	2-methylpentene
23	n-hexane
24	methylcyclopentane
25	2,4-dimethylpentane
26	benzene
27	cyclohexane
28	2-methylhexane
29	2,2-dimethylbutane
30	3-methylhexane
31	2,2,4-trimethylpentane
32	n-heptane
33	methylcyclohexane
34	2,3,4-trimethylpentane
35	toluene
36	2-methylheptane
37	3-methylheptane
38	n-octane
39	ethylbenzene
40	m-xylene
41	p-xylene
42	styrene
43	o-xylene
44	n-nonane
45	isopropylbenzene
46	n-propylbenzene
47	1,3,5-trimethylbenzene
48	1,2,4-trimethylbenzene
49	o-ethyltoluene
50	m-ethyltoluene
51	p-ethyltoluene
52	1,4-diethylbenzene (para)
53	1,2,3-trimethylbenzene
54	n-decane
55	n-undecane

- Prepare ratios of individual species comparisons.

Hydrocarbon species measured in the ambient air have anthropogenic and biogenic origins and can be the products of photochemical reactions. Since some species' reaction rates are significantly slower than others and these species are predominately associated with certain source types, they can be used as tracers of those sources. Thus, species ratios can be used to determine dominant source categories. Investigating ratios of individual chemical species may help to identify the presence of unique regional sources for a species at a particular monitoring site. Because mobile sources are a major contributor to hydrocarbon emissions, it is useful to examine ratios of ambient hydrocarbons that are characteristic of motor vehicles and to compare them to the ratios of the same species reported in the emission inventory. Ratios commonly used to identify mobile source emissions include acetylene/benzene, toluene/benzene, xylene/benzene, ethylene/acetylene, CO/benzene, and CO/acetylene (Main et al., 1999).

Example Ratio Analysis

Site	Acetylene/ benzene	Toluene/ benzene	Xylene/ benzene	Ethylene/ acetylene	CO/benzene	CO/acetylene
Ambient - Site 1	**1.6**	**4.6**	**3.3**	**1.2**	**145**	**92**
Emission Inventory – All Sources	1.0	4.7	0.6	2.3	157	154
Emission Inventory – Mobile Sources	1.1	2.8	0.6	2.0	151	141
Ambient - Site 2	**1.0**	**3.3**	**2.6**	**2.0**	**224**	**227**
Emission Inventory – All Sources	1.2	3.0	0.5	2.6	126	108
Emission Inventory – Mobile Sources	1.2	2.8	0.6	1.9	163	134
Ambient - Site 3	**1.5**	**3.6**	**3.3**	**1.2**	**N/A**	**N/A**
Emission Inventory – All Sources	1.1	4.5	0.7	2.2	136	123
Emission Inventory – Mobile Sources	1.1	2.7	0.6	2.1	151	131
Ambient - Site 4	**0.5**	**3.9**	**2.9**	**2.6**	**168**	**333**
Emission Inventory – All Sources	1.1	4.4	0.7	2.3	168	156
Emission Inventory – Mobile Sources	1.1	2.7	0.6	2.1	158	143
Tunnel	**1.9**	**2.5**	**2.3**	**N/A**	**N/A**	**N/A**

Data are 0500, 0600, 0700 ST.

Emission inventory evaluations should be conducted as part of an iterative process:

Step 1: Prepare the emission inventory.

Step 2: Evaluate the emission inventory using a top-down approach.

Step 3: Revise the emission inventory using bottom-up improvements (i.e., using new/improved emission factors, activity data, temporal and spatial allocation, speciation profiles).

Step 4: Reevaluate the emission inventory using a top-down approach.

Top-down Emission Inventory Evaluation

Strengths

- Provides a method to assess parts of an emission inventory that appear to be suspect; improvements can be made prior to photochemical modeling.

- Can assess detailed chemical species composition between the inventory and ambient air.

- Can greatly improve emission estimates.

Limitations and Uncertainties

- Requires extensive data.
- Uncertainties in the emission estimates affect comparisons.
- Uncertainties in the ambient data measurements affect comparisons.
- Requires ambient speciated data.
- Assumes that the emission inventory NO_x estimates are reasonable.

Comparison-related uncertainties include:
- Proper temporal and spatial matching of emissions data and ambient data.
- Meteorological factors.
- Level of ambient background concentrations and chemical reactions.

Receptor models provide empirical relationships between ambient data at a receptor and emissions by source category. The fundamental principal of receptor modeling is that mass conversion is assumed and a mass balance analysis is used to identify and apportion sources of ozone precursors in the atmosphere. Receptor models are useful for resolving the composition of ambient primary VOCs into components related to emission sources.

Three main types of receptor models:

- Models that apportion primary VOCs using source information.
- Models that apportion primary VOCs without using source information.
- Models that apportion primary and secondary VOCs.

> There are more than a dozen currently existing receptor models; however, EPA's OAQPS has only recognized the chemical mass balance model and principal component analysis as part of its State Implementation Plan development guidance documents.

Strengths

- CMB accepts inputs and creates outputs in a wide variety of formats.

- PCA is widely available in statistical software and relatively easy to perform.

- PCA does not require source profiles.

- CMB generates errors in source compositions accepted by OAQPS.

- CMB and PCA have been accepted by EPA's OAQPS for SIP development.
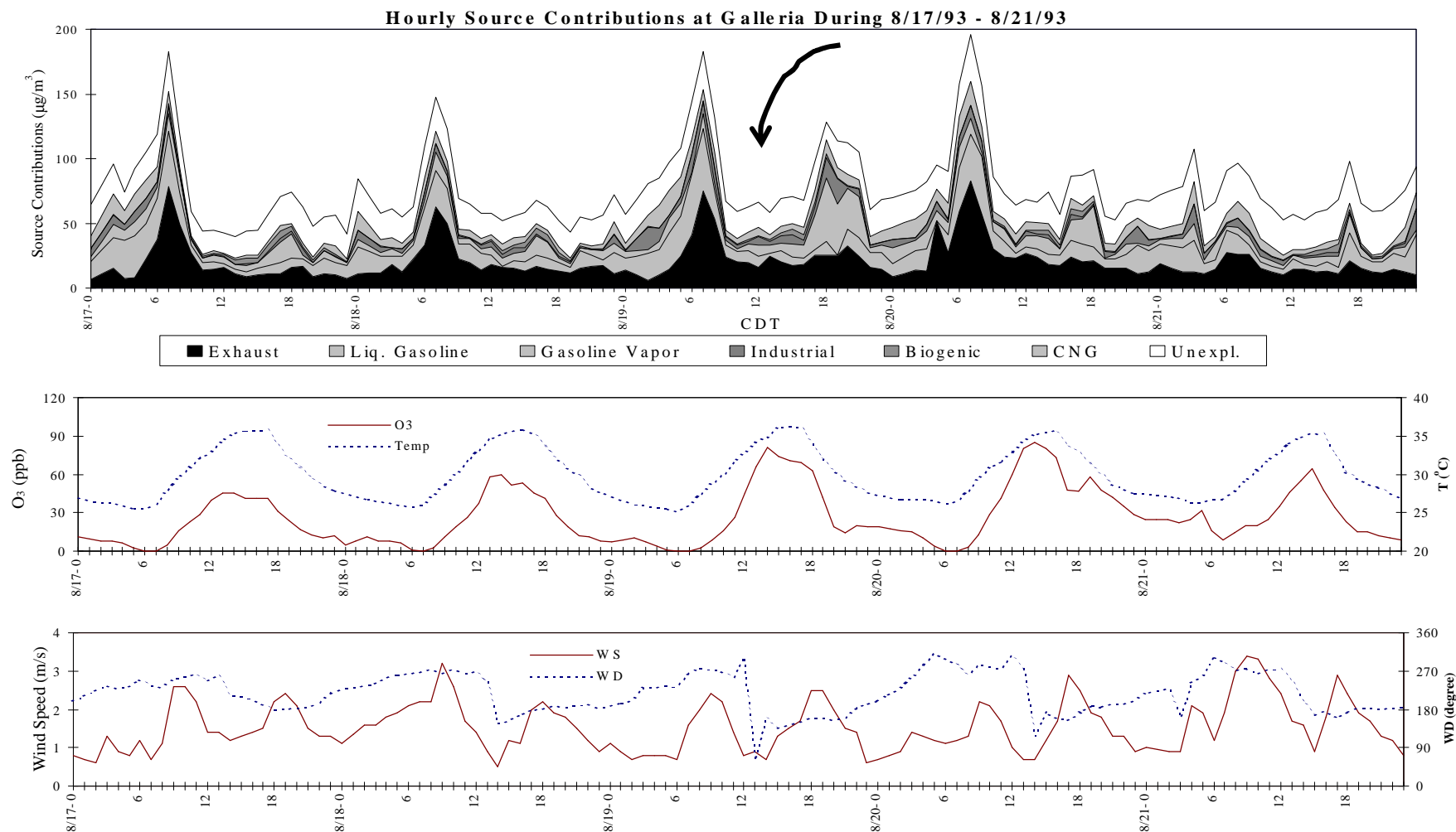
Limitations

- CMB requires source profile information.

- PCA requires the analyst to infer which sources are contributing.

- Results of the CMB model are only as accurate as the speciation profile input data.

- CMB can mis-specify emission sources.

- CMB is sensitive to collinearity.

Mobile Source
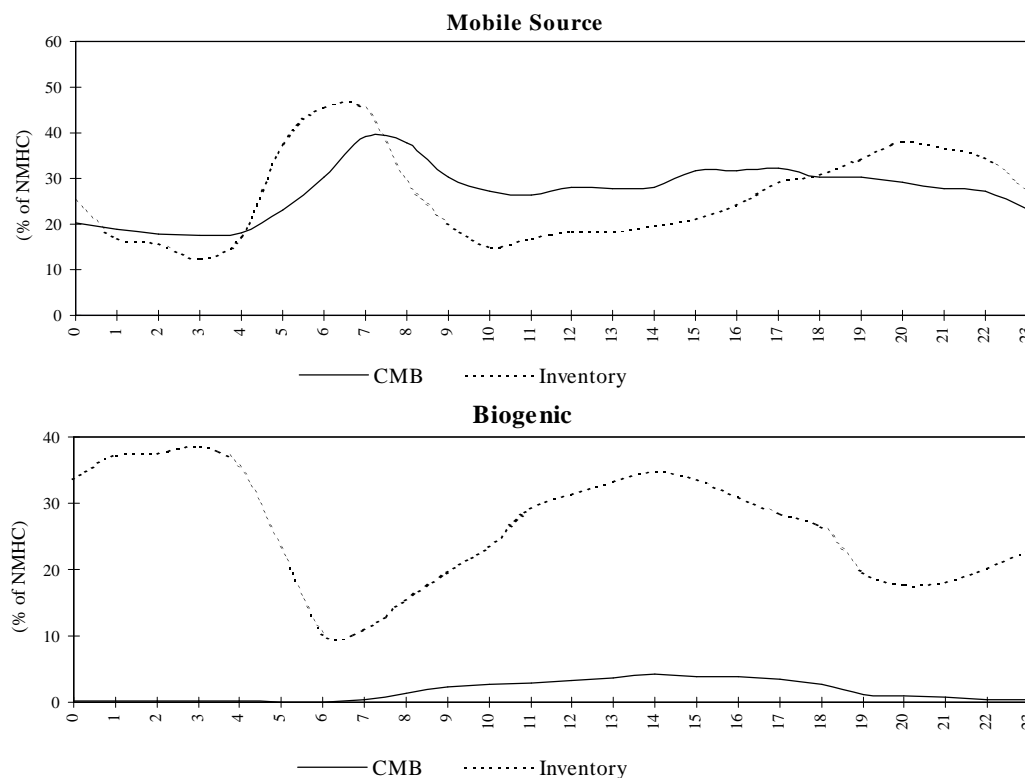
Biogenic

- Comparisons between CMB results and the emission inventory for August 19, 1993 at Galleria (Houston), TX (Lu and Fujita, 1995).

- For mobile sources, the inventory shows an earlier maximum contribution than the CMB results indicate.

- For biogenic sources, the CMB results are a lower limit of ambient biogenic contributions because isoprene is the only measured biogenic VOC. The nighttime biogenic contribution indicated by the inventory appears odd.

Hourly Source Contributions at Galleria During 8/17/93 - 8/21/93

Legend: ■ Exhaust ▨ Liq. Gasoline ▨ Gasoline Vapor ▨ Industrial ▨ Biogenic ▨ CNG ☐ Unexpl.

Hourly source contributions, ozone concentrations, temperature, wind speed, and wind direction at Galleria (Houston), TX during August 17-21, 1993 (Lu and Fujita, 1995). Note that on August 19, the wind shifted resulting in a greater gasoline vapor contribution consistent with sources in that direction.

Available Methods and Data (1 of 3)

- Emission inventory data sources:
  - State and local air quality management agencies
  - EPA National Emissions Trends Inventory:
    http://www.epa.gov/ttn/chief/ei/
  - Emission inventory improvement program guidance documents:
    http://www.epa.gov/ttn/chief/eiip/techrep.htm

- Emissions processing software:
  - EMS95: see *www.ladco.org*
  - SMOKE: see *http://envpro.ncsc.org/products/smoke/*

- Ambient data sources:
  - AIRS Data via public web site: *http://www.epa.gov/airsdata*
  - AIRS AQS via registered users: register with EPA/NCC (703-487-4630)

- Source profile sources:
  - EPA SPECIATE (*http://www.epa.gov/ttn/chief/software.html#speciate*) was recently updated and now contains many measurements conducted as part of recent studies (e.g., NARSTO-Northeast) including:
    - Callahan tunnel in Boston (MA), Lincoln tunnel (NY), Fort McHenry tunnel (MD), Van Nuys and Sepulveda tunnels (CA), Tuscarora tunnel (PA), Federal building garage (MA) all from 1995.
    - Auto-Oil Program measurements for 1989 and 1983-1985 fleets.
    - Gasoline liquid and/or vapor composites from Boston, Los Angles, Seattle, and El Paso (1995 and 1996).
    - Degreasing and solvent use, consumer products, industrial sources (i.e., refineries, chemical facilities), oil and gas production, burning, and surface coatings.

Available Methods and Data

- Source profile sources (continued):
  - Literature review: Additional on-road vehicle exhaust profiles have been developed from measurements in the San Francisco Bay Area, Caldecott Tunnel (Kirchstetter et al., 1999), 1993 Texas source profiles (Fujita, 1995), and elsewhere.
  - Federal Test Procedure measurements for 1975-1982 model years (Sigsby et al., 1987).
  - Local, state, and federal agencies; for example, California has information available at *http://arbis.arb.ca.gov/emisinv/emsmain/emsmain.htm.*
  - Analysis of ambient data (e.g., using UNMIX, PMF).
- Meteorology data sources:
  - Meteorological parameters from NWS: *http://www.nws.noaa.gov*
  - Meteorological parameters from PAMS/AIRS AQS: register with EPA/NCC (703-487-4630)
  - Private meteorological agencies (e.g., forestry service, agricultural monitoring, industrial facilities)
  - State and local agencies

Issues Associated with Emissions Evaluations Using Ambient Data

Ambient air quality data can be used to evaluate emission estimates and source apportionment; however, the following issues should be considered:

- Proper spatial and temporal matching of emission estimates and ambient data.

- Proper matching of measur<u>ed</u> VOCs with measur<u>able</u> VOCs in the inventory.

- Ambient levels of background NO_x, CO, and VOC.

- Meteorological effects on the comparison.

Caution: *Ambient/emission inventory comparisons are useful for examining the relative composition of emission inventories; they are not useful for verifying absolute amounts unless they are combined with bottom-up evaluations.*

Summary

- Emission inventory evaluation is an important part of the PAMS program. A high quality emission inventory is needed to best support air quality modeling, exposure modeling, and control strategy development.

- A variety of evaluation techniques are available including engineering judgment, "bottom-up" evaluations, "top-down" evaluations, and receptor modeling.

- PAMS data are a useful part of an emission inventory evaluation and improvement process.

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